

## CENTRIFUGAL REACTOR WITH RESIDENCE TIME CONTROL

This invention relates to a fluid control device for chemical processing. The invention has application to chemical reactors and in particular to a  
5 method and apparatus employing thin film technology.

All chemical reactions involve changing the molecules involved. Reactive components can present themselves as liquids, solids, gases or vapours. A perfect reactor would provide the ideal pattern of contacting for an  
10 optimal time to achieve the desired outcome.

Most reactions in the laboratory and in batch production involving liquids utilise stirred or agitated vessels to provide the necessary contacting between components. The liquid phase may hold a number of the reacting  
15 species in solution and molecular contact. Unless impeded by micelles, this contact is normally intimate and mainly impeded by viscosity.

One or more of the components in the liquid phase may be immiscible and may take the form of an emulsion where molecules in the suspended  
20 phases contact the components in the continuous phase and any other immiscible components by diffusion. Similarly gases may be present in the form of bubbles.

In most cases the more severe the agitation the faster the reaction  
25 proceeds until it is only limited by the kinetics of the chemistry. The reaction proceeds with time until it is deemed complete. Time proceeds at the same rate throughout the tank and so as long as the agitation level is evenly spread at a more than necessary level throughout the tank, the reactions are evenly spread through the reaction vessel with reaction  
30 progressing evenly throughout the vessel with time.

Immiscible components are those in which the molecule population density of the other components is substantially lower than could be achieved were the components fully soluble in each other. Thus, solids, gases and vapours may be described as immiscible components as they are  
5 substantially immiscible with the liquid components.

It is recognised that often process outcomes depend on diffusion between and into these media and so mixing and diffusion path lengths are important aspects of performance.

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Difficulty is encountered with traditional batch reaction equipment because the reactions occur at a molecular scale and the mixing is on a much greater scale. On scale-up from the laboratory it is increasingly difficult to maintain the ideal reaction conditions throughout the reaction  
15 space.

For example, transferring heat into and away from energetic reactions can prove difficult. Bulk mixing is generally easy but mixing at a microscopic level is difficult to achieve and discontinuous phases tend to  
20 aggregate so bubbles become larger and liquids of different density separate.

All of these factors, and more, severely limit the effectiveness of reactors and so yields and adverse by-products can be far from ideal. Molecular  
25 mobility can be enhanced by lowering viscosity and so many reactions are often conducted utilising solvent systems to lower the viscosity and improve miscibility.

Much of chemical engineering effort has been directed to countering these effects on scale-up and chemists, being aware of the difficulties of scale-up,  
30 up, normally endeavour to provide process sequences which minimise

these known difficulties. Solvents in particular are utilised to bring reactive components together and to separate out products both desirable and undesirable. A less scale sensitive reaction sequence can be achieved by deliberately slowing the reactions by diluting the components, limiting the feed rates and operating at low temperatures.

It is well known that thin films can be utilised to provide an intimate contact with a heat transfer or catalysed surfaces and that disturbing the film can assist mixing and refresh the surface to promote mass transfer with a gas or vapour.

Such films can be created for example by wiping and spreading and there are many generic devices to achieve this outcome. Providing a packed structure that permits liquid components to drip from one packing component to the next are known to improve the effectiveness of liquid/gas mass transfer.

The present invention has been made from a consideration of the foregoing and is more fully described hereinafter with reference to Figures 1 to 16 of the accompanying drawings that depict various features according to different aspects of the invention that may be employed separately or in combination as desired.

The liquids referred to in this application may be single components, multiple components in solution, immiscible combinations of liquids, liquids containing non-liquid components, and solids that are able to flow like liquids, such as spherical beads or powders or solids suspended on or in liquids. The liquids may carry with them homogeneous or heterogeneous catalysts including phase transfer catalysts.

According to one aspect, the present invention uses means such as perforations or edge features, for example notches, in an inclined supporting plane carrying liquid in an enhanced gravitational field to produce fibrils of liquid which can be utilised to promote mixing, enable  
5 free gas and vapour passage

The perforations and/or edge features (see Figs 2 and 3) can be used to provide different residence times for the molecules flowing through the equipment to provide beneficial material properties.

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The perforations and/or edge features may be used on inclined planes to provide for controlled mixing of a number of liquids to provide a closely controlled chemical reaction sequence.

15 According to another aspect, the present invention utilises gravitational forces, normally induced by centripetal acceleration, to facilitate the generation of thin films.

The liquid when unsupported falls freely in the field until it encounters an  
20 accelerating element (see Fig. 1). If that element is large enough and inclined to the acceleration field the liquid will spread and form a thin film the depth of which can generally be defined by Nusselt's equation for flow down an inclined plane.

25 According to yet another aspect, the present invention uses fibrils induced by flowing the film through holes, cuts and notches (see Figs. 2 & 3) that can be utilised to provide for controlled residence time distribution, diffusion, mixing and reactions.

30 Even when utilising substantially immiscible components, a reactive environment is provided in which the components can be brought into

intimate contact with each other in a flowing stream or streams. The time taken for the reaction to proceed also involves a displacement through the reactor.

- 5 According to a still further aspect, the present invention uses a combination of immiscible liquids flowing on an inclined plane which may be subjected to enhanced gravitational fields to produce a flow close to plug flow at Reynolds numbers which are very small where normally the flow profile of a single component would normally be a parabolic  
10 laminar flow profile with zero velocity relative to the plane as its minimum.

Embodiments of the invention permit fluids to achieve much thinner films than could be achieved with the same equipment and with the same mass  
15 flow rate with any one of the fluids flowing on its own.

Chemical reactants including liquids, solids and vapours can be induced to pass readily between the films enhanced by short path lengths and gravitational forces where there is a density difference.

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Temperature control can be facilitated by this vapour transport by evaporation and condensation

Embodiments of the invention also describe how the streams may be  
25 selectively separated and re-combined by utilising troughs in the planes to gravitationally separate out components that may be re-introduced later.

As well as conventional reactions, the use of more than one immiscible liquid provides the opportunity to utilise one or more liquid electrodes to  
30 create an electrochemical cell which may have plate separations of a few microns.

Ideally, some embodiments of the invention seek to achieve a flow field where the flow is entirely one dimensional (plug flow) with the distance through the reactor being a function of the time that the reactants have  
5 been present within the reactor irrespective of the position of the molecules in the film cross-section. The more time sensitive the reaction, for example competitive reactions where the products react with themselves and/or the feedstocks, the more critical it is that plug flow should be achieved.

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It is normally assumed that the best characteristic that can be obtained for a flow reactor is limited by the velocity profile, with a laminar flow profile being a typical representation of the residence time distribution. This is normal for a reactor operating under high acceleration where one  
15 normally changes the film shear, film thickness and residence time by modifying reactor rotational speed and the stream massflows. However, the present invention provides other methods to control these which can be used to improve the outcomes of processes.

20 More particularly, some embodiments utilise combinations of immiscible films in acceleration fields which may be substantially greater than that of gravity resulting in a technique that permits the residence time profile of one or more of the liquid components to undergo substantially plug flow (see Figs. 4,5 & 6) in a film with a Reynolds number well below 100 and  
25 more often well below 1.

The films can be maintained with a thickness of between 1 and 1000 microns in many cases. The film profiles are affected by the fluid density, mass flow and viscosity as well as the geometry and acceleration.

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Fluids can be chosen to achieve a specific profile, for example if a low viscosity liquid flows down an inclined plane together with a higher viscosity, lower density, immiscible material e.g. liquid metal and polymer, then the result is that the low density material will form a  
5 separate layer above the high density material.

The liquid next to the supporting plane will be substantially stationary with respect to it but the surface away from the supporting plane will be relatively free flowing in the direction of the slope. For any given  
10 acceleration the thicker the first film and the lower its viscosity the faster its surface will flow.

The higher viscosity material which is subjected to the same acceleration will move much more readily than it would have done were it contacting a  
15 solid wall and hence be much thinner for a given mass flow and have a relatively small difference in velocity between the bottom and the top of the film.

Thus, in this circumstance not only is the flow close to a plug flow for  
20 the film away from the surface but the film thickness for a given flow is much thinner than it would otherwise be. This reduces residence time, the residence time distribution and diffusion path lengths all of which may be beneficial.

25 In the case where the reaction is significantly exothermic or endothermic, it is possible to utilise evaporation or condensation from or to one or more of the liquid streams (see Figs. 7 & 8). This may be done as well as utilising the heat capacity of the flowing liquids, heat exchange to the supporting planes, energy exchange by infra-red, radio, ultra-violet,  
30 inductive, light and vibration, and to the gas or vapour flowing above the free surface.

It is also possible to introduce an immiscible stream specifically intended to provide thermal control by evaporation or condensation. Where vapour exchange is occurring on the free surface, temperature control can be extremely tight as vapour exchange can be at very high rates and the pressure closely controlled.

Where a volatile film is located in one of the lower streams, it may evaporate by nucleate boiling but more commonly will undergo evaporation and hence flash cooling when it falls from one plane to the next. The heat of reaction in the liquid on the supporting plane may be absorbed as sensible heat in this and all the other components as they flow down the plane and may be released in part or in whole in vapour exchange, with a consequential temperature change, as the components fall from one plane to the next.

Where the reaction is exothermic or endothermic the density change produced by the heat of reaction may be utilised to move the reacted components away from the reaction site (see Fig. 9).

Given the low Reynolds number encountered when such films are flowing down inclined planes it is difficult to induce effective mixing by perturbation. Under the effects of high acceleration fields, when the fluid leaves the surface it tends to form a thin sheet, as the surface accelerates away from it, with a thickness substantially less than that of the film on the supporting plane.

We have found however that, if the liquid is permitted to flow through holes in the supporting plane or over notches in the edge it can be induced to form fibrils rather than sheets (see Figs 1, 2 & 3). As the distance from its point of origin increases the fibril, of a non-fibre forming



material, will ultimately coalesce into droplets of a similar scale to that of the fibril.

5 This fibre forming effect substantially changes the flow profile of the flowing film leaving the plane, increases the surface to volume ratio of the liquid and so ensures both good mixing and surface renewal when passing from one supporting plane to the next.

10 The mixing effectiveness can be further enhanced by inducing a velocity difference between the one plane and the next (See Fig 10). This can be further enhanced by features on the receiving plane impacting on the falling fibrils and droplets causing them to break up.

15 In equipment generating high acceleration fields, with significant gas velocities, to deliberately form fibrils has the advantage of not impeding gas flow as much as sheets of liquid. This can be critical as there can be a significant pressure build up from one side of the sheet to the other which can force the film to be displaced or broken up randomly or even to substantially impede the gas flow. This latter effect is particularly  
20 critical with low pressure systems such as are routinely required in devolatilisation devices or polycondensation reactors.

25 Gases and vapours can be induced to flow radially inwards if so desired by the use of appropriate seals. These may be liquid seals. Such seals can also be utilised to provide for different chambers in the reactor which may contain gases or vapours of different compositions and/or pressures.

30 Because the films are substantially laminar there is excellent micro-mixing along the shear plane which promotes contact between species in that plane and hence reaction. Mixing normal to the shear planes is

achieved by diffusion and the short path length provided by the thin films promotes this.

5 The novel techniques described above provide for ways of thinning the films and provide a controlled shear in the reacting film. We have discovered that it is advantageous to repeatedly break the film into fibrils and then re-form them into a rectilinear flow field on a subsequent surface. Increasing the frequency of this action increases the mixing.

10 This effect is particularly important when solubility or diffusion rates are limited, for example for viscous polymers requiring to be stripped of a volatile component. Also the dispersion of fine solids in the liquid stream is enhanced as the buoyancy forces due to density differences are substantially eliminated in the free-falling fibrils.

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The desired frequency of these jumps from one surface to another may be chosen according to the requirements of the process. For example, if a process is dominated by mixing solid particles into a liquid stream then, in general, the more frequent the interruptions should be. Where there is  
20 little requirement for frequent surface renewal and general mixing, then the more the surface can beneficially be continuous.

Cascading supporting planes can be in the form of a series of stepped jumps in the plane such as one would see in a series of waterfalls. They  
25 can also be nested, coherent or random separations between planes. In the limit, they can be of a similar scale to the fibrils and be positioned to simply break the fall of the fibrils and re-form them using an ordered or random structure.

30 In some circumstances, there may be a requirement for a reactor not to show a plug flow characteristic. For example, in polymerisation

reactions, such as ring opening polymerisation, as long as un-connected rings are available a catalysed polymer chain will keep growing. Thus a true plug flow reactor would produce a very narrow molecular weight distribution. If a wider molecular weight spread is required or, in the limit, a particular population of different sized molecules, then there is a requirement to provide different residence times for the population of molecules. An ideal reactor would achieve this residence time distribution by design.

This can be readily achieved with the present invention by utilising a reactor format where some of the liquid flow falls onto a plane below it in the acceleration field earlier than other parts of the stream.

Thus, for example, inclined planes may be nested (see Fig 11) such that the first plane is sloped one way and the second plane is sloped the other way under the first plane with a hole made in the first plane part way down the slope. Alternatively or additionally, part of the first plane may have a shorter path length. As a result, some liquid will exit onto the second plane sooner than other parts of the flow stream thus avoiding the requirement to flow the full length of the first plane and from the start of the second plane.

The same effect could be achieved by having some of the liquid on planes in higher or lower acceleration fields, of different lengths, number of planes, of different slopes or any combination of these.

Another embodiment uses blading in a spiral form (see Fig. 13) which provides a continuous slope into which holes, notches or features may be cut to achieve residence time control.

The operating principles above described can be readily employed in a rotating reactor operating at an acceleration (acceleration due to rotation = speed of rotation<sup>2</sup> x radius) well in excess of the earth's gravitational field with the liquid stream supporting planes of the reactor inclined with respect to the resulting acceleration field.

The planes may be in the form of cones, truncated cones (see Fig 12), spirals or coherent or random packings. The inclination angle of the planes may vary from being in line with the acceleration field to normal to with constant angles, discretely changing angles or curved forms. It is possible for the flow to continue with the acceleration field reversed utilising surface tension to hold the fluid to the surface. This situation is encountered for example where the surface of a tube is used with the axis of the tube normal to the acceleration field. Where the angle approaches that of the acceleration field, the flow may change from laminar to wavy. This may beneficially enhance the mixing and surface renewal effects.

Further to this residence time control, it is possible create very thin coherent films of different density immiscible liquids and solids. These may be thin enough to substantially overcome the diffusion barriers which would otherwise substantially limit the rate of chemical reaction.

Whenever there is a density difference between materials an enhanced acceleration field will increase buoyancy and so tend to separate those components. Thus, components that normally would not separate readily, and emulsions, will tend re-form into coherent thin films.

Immiscible reaction products, whether solid or immiscible liquid, can be separated readily from the reactants if there is a density difference between them and the reactants. They may be carried preferentially into other streams to be dissolved or be swept along with a stream or be

carried between two liquid streams of greater and lesser density or to the surface or the supporting plane.

5 The highest density component can be separated from the others in the stream by flowing it into a well (see Fig 14). If a sequence of such wells is presented to the flow stream then all of the different density streams can be extracted and removed or re-formed in any combination and sequence. The outflow from the well can be achieved via a weir as shown or by any other means such as a float valve or a pressure operated  
10 valve.

The use of nested inclined planes also facilitates the introduction of new components to the flow stream in a controlled way. For example, if it is desired to add a new component slowly to the stream, it can be selected to  
15 flow through a hole(s), notche(s) or edge(s) in a first feed plane positioned such that the flow encounters a flow on a second plane at different points down the incline. If these two planes are conical in form and rotating at different speeds, then each feed flow would be delivered evenly parallel to the direction of flow on the second cone.

20 Figure 15 shows two nested cones where a liquid can be introduced in Zone 1. Another liquid may be added in Zone 2 to exit at a single point along the cone if holes are located by an annular barrier. Zone 3 shows liquid added continuously over a length of the zone. Zones 4 and 5 show  
25 liquid added in a controlled sequence from two or more discrete streams. Zone 6 and 7 show liquid added firstly over a length of the cone from Zone 6 and subsequently from Zone 7.

One advantage of such a device is that the film distribution characteristics  
30 of the cone can be utilised to evenly distribute the relatively stable feed materials before they encounter the instability inherent within a reactive

environment. Feed materials may also be fed into the system and removed utilising troughs and tubes.

If such a feed cone is segmented then number of different materials can  
5 be added in a controlled sequence. Some of these materials may free flow  
or be pumped from the separated process streams or be new materials.  
Pumping may utilise the peristaltic pressure available from the fluid itself  
and/or other means. The use of pumping allows for the possibility of one  
10 more of the liquid or solid components being moved radially inwards in  
stages thus allowing for a stepwise counter-flow of liquid and solid  
components against the acceleration field.

The flow on the feed cone itself may be part of the reactive sequence such  
as activating feed materials immediately prior to joining the main reaction  
15 sequence. This may require a further feed cone inboard of the first one  
and so on. It is possible for there to be a significant number of feed  
systems located radially inboard of each other, axially displaced or close  
coupled as separate devices.

20 It is also possible to enhance reaction utilising heterogeneous or  
homogenous catalysts fixed or flowing with the streams. It is also  
possible to utilise ultra-violet light or other energising field radiating  
through the films or fibrils. Where critically timed exposure is required  
the radiation source can be incorporated into the supporting plane or  
25 masked by the addition or removal of an opaque film.

It is also possible for electric fields or currents to be applied to the  
streams to promote electrochemical reaction (see Fig. 16). It may be  
necessary in such devices to have insulating solid materials and non-  
30 conducting fluids introduced to facilitate this. Where significant currents  
may flow or to facilitate application of the electrical fields, electrodes

may be introduced into one or more troughs in the inclined plane to facilitate effective coupling to a liquid stream. Thus conducting liquid streams may in themselves form electrodes which may or may not produce reactive species. It is possible to induce the required currents by  
5 inductive coupling utilising a magnetic field or by capacitive coupling through the films.

According to another aspect, the present invention provides a chemical reactor embodying any one or more of the novel features described herein  
10 separately or in combination.

Examples of two reactors embodying the invention are illustrated in Figures 17 and 18 where Figure 17 shows a co-rotating laboratory reactor with evenly spaced holes (not shown) along the edge of each blade and  
15 Figure 18 shows a co-/counter-rotating reactor within a pressure controlled shell.

As will now be appreciated from the foregoing description of exemplary embodiments, the present invention utilises the characteristics of flow in  
20 an enhanced acceleration field to provide a reaction environment able to be tuned to be close to optimal that can be employed in a number of chemical systems.

It is a general requirement for the reactants entering a reaction system to  
25 be sufficiently stable to endure the time required to complete a controlled reaction. The reactor is deliberately intended to reduce the feedstock stability by catalytic or chemical means and convert the reactants into desired products with high effectiveness. An ideal reactor would therefore take in stable reactants, promote the desired instability and re-  
30 stabilise conditions before exit. Ideally a series of reactive steps could be

taken in a single device. The spinning cone reactor of the present invention may be constructed to achieve this objective.

As will be apparent, the present invention provides a method and apparatus for effecting chemical reactions employing thin film technology having numerous benefits and advantages that can be used in a spinning cone reactor as described.

For example, by using notches and other edge features, and perforations in an inclined supporting plane carrying liquid in an enhanced gravitational field, fibrils of liquid are produced which can be utilised to promote mixing, enable free gas and vapour passage. Furthermore, these perforations and edge features can be used to provide different residence times for the molecules flowing through the equipment to provide beneficial material properties. Moreover, these perforations and edge features on inclined planes can be used to provide for controlled mixing of a number of liquids to provide a closely controlled chemical reaction sequence.

It will be understood that the exemplary embodiments described herein are intended to illustrate the diverse range and application of the invention and that features of the embodiments may be employed separately or in combination with any other features of the same or different embodiments.

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Moreover, while the exemplary embodiments described and illustrated are believed to represent the best means currently known to the applicant, it will be understood that the invention is not limited thereto and that various modifications and improvements can be made within the spirit and scope of the invention as generally described herein.

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